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E. K. Hulet, J. F. Wild, R. W. Lougheed,
P. A. Baisden, R. J. Dougan, and M. G. Mustafa
Lawrence Livermore National Laboratory
Livermore, California 94550

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Lawrence Livermore National Laboratory, Livermore, California 94550, USA

Abstract - The fragment energies of about 725 coincidence events have now been observed in the spontaneous fission (SF) decay of 105-min ^{259}Md since its discovery in 1977. The fission of ^{259}Md is characterized by a symmetric mass distribution, similar to those of ^{258}Fm and ^{259}Fm, but with a broad total kinetic energy (TKE) distribution which peaks at about 195 MeV, in contrast to those of ^{258}Fm and ^{259}Fm, for which the TKE is about 240 MeV. We have postulated that this kinetic energy deficit, ~40 MeV, is due to the emission of hydrogen-like particles by ^{259}Md at the scission point in a large fraction of the fissions, leaving the residual fissioning nucleus with 100 protons. The residual nucleus would then be able to divide into two ultrastable tin-like fission fragments, but with less kinetic energy than that observed in the SF of ^{258}Fm and ^{259}Fm, due to binding-energy losses and a reduction in the Coulomb repulsion of the major fragments. To test this hypothesis, we have performed counter-telescope experiments aimed at detecting and identifying these light particles. In 439 SF events we have observed 3 ± 3 protons of the appropriate energy, too few to account for the kinetic energy deficit in the fission of ^{259}Md. There seems to be no explanation for this problem within the framework of current fission theory. These results are discussed along with preliminary measurements of light-particle emission in the SF of ^{256}Fm.

Following our discovery of the SF-decay nuclide ^{259}Md, we performed experiments to deduce the fission properties of this isotope by observation of the kinetic energies of coincident fission fragments (Ref. 1). The resulting mass distribution was quite symmetric, similar to those of the SF of ^{258}Fm and ^{259}Fm (Ref. 2,3). However, the total kinetic energy (TKE) distribution, especially at mass symmetry, was 40 MeV lower than for ^{258}Fm and ^{259}Fm. The ^{259}Md TKE is most comparable to those of ^{256}Fm and ^{257}Fm. Such a low TKE associated with symmetric mass division is unusual and is inconsistent with current fission theory in which fragment shells appear to govern the fission process. Symmetric division of the heavy Fm isotopes leads to fragments approaching the magic nucleon numbers Z = 50, N = 82 which, due to their spherical rigidity, possess low deformation and internal excitation energy. Therefore, fissions with near-symmetric mass division exhibit correspondingly higher TKE's than those with asymmetric division, which yields fragments that are soft toward deformation.

Two possible explanations have been proposed to account for the kinetic energy deficit in ^{259}Md: (a) two-body fission with either or both fragments highly deformed; (b) three-body fission, i.e., two-body fission accompanied by the emission of a light particle (Ref. 1). Studies based on Strutinsky shell corrections from the two-center shell model do not show a stable minimum in that region of the potential energy surface which would render the first explanation valid. The only stable minimum, according to this prescription, corresponds to two nearly spherical fragments.

Therefore, we decided to test the more likely explanation, the emission of a light particle accompanying fission into two nearly identical, spherical fragments. This explanation for the high symmetry of the mass distribution seemed highly plausible since the emission of a Z = 1 particle from a Z = 101 nucleus at scission would leave the residual fissioning nucleus with 100 protons, resulting in a symmetric mass distribution similar to those of ^{258}Fm and ^{259}Fm. The lowering of the fragment energies, especially for symmetric fission, arises from the disturbance of the Coulomb field of the two major fragments by the charged particle between them.

Two similar counter telescopes were constructed, each consisting of a ΔE detector and an E detector placed above the sample, and a conventional Si surface-barrier detector below it. A coincidence (2τ = 470 ns) was required between the ΔE and E detectors as evidence of a light particle; the ΔE detector measured energies between 0.75 and 15 MeV, while the E detector measured energies from 0.5 to 30 MeV. For a valid light particle, the energies obtained from a ΔE-E coincident event were required to fall within certain limits based on known range-energy relationships of light particles for Si. In addition to light particles, fission-fragment kinetic energies were also obtained during these experiments by analyzing coincident fission pulses from the detector below the sample and the ΔE detector above.

The detector systems were calibrated with sources of ^{252}Cf evaporated from an aqueous solution onto thin VYNS films. The light-particle calibration data are shown in Fig. 1 for both systems as plots of the energy measured by the ΔE detector vs. the total particle energy. The envelopes shown for each type of particle represent the range of calculated energies which should be deposited in the ΔE detector for a given kind of particle, its incident energy, and the angle at which it entered the detector. The ratio between the number of long-range alpha particles and the number of fissions recorded by the fission detector below the sample provided a calibration with which to compare our ^{259}Md light-particle results.

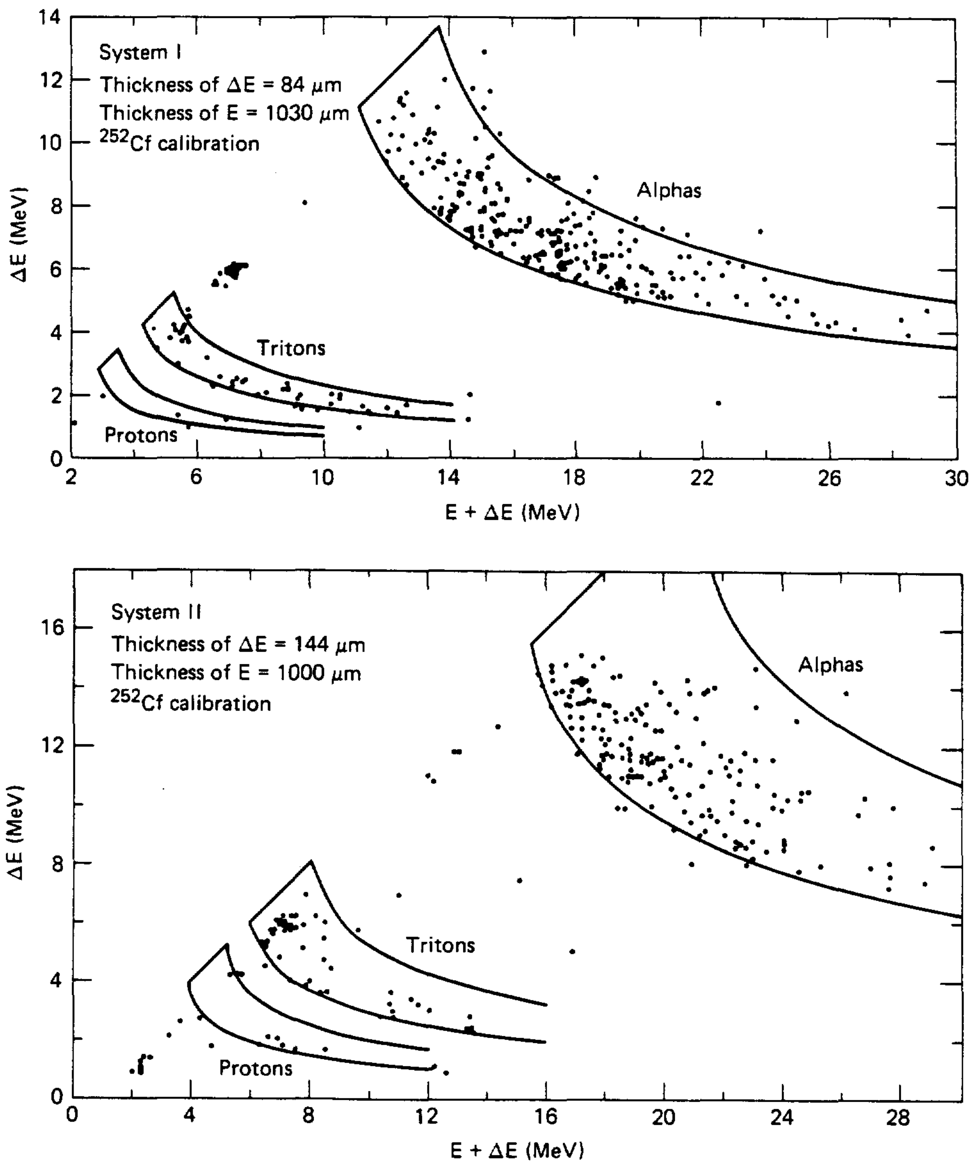

Fig. 1. Energy deposited in the ΔE detector vs. total particle energy for the ^{252}Cf calibration of the two counter telescopes used in these experiments.

Samples of ^{259}Md were prepared by chemically separating 60-min ^{259}No from the recoil products of the bombardment of ^{248}Cm with 96-MeV ^{18}O ions. The ^{259}No was evaporated from an aqueous solution onto thin VYNS film, then positioned in the counter telescope inside an evacuated chamber. The ^{259}No decays by electron capture to ^{259}Md with a 25% branching ratio, thus serving as a radiochemically pure source of ^{259}Md SF. Samples from eleven bombardments were counted for a total of over 4800 minutes in the two systems; in addition, background counts over 17,450 minutes long were taken in each system. All of the events detected in both the ^{259}Md light-particle runs and the background counts are shown in Fig. 2 as plots of ΔE vs. E_T.

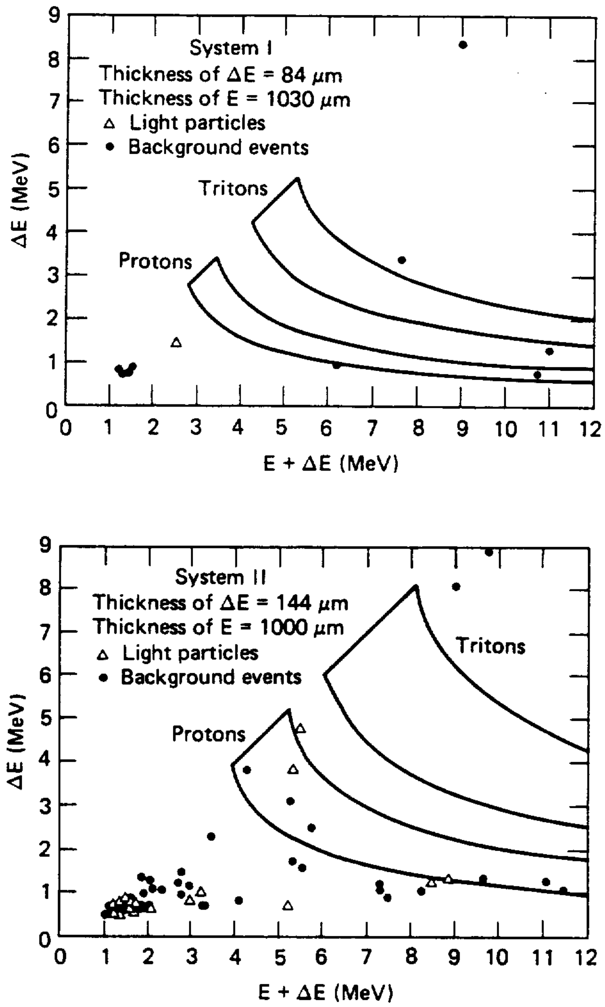

Fig. 2. Energy deposited in the ΔE detector vs. total particle energy for the light-particle events detected from eleven samples of ^{259}Md and also the events obtained during over 17000 min of counting background.

A careful analysis of these results yields a value of 1.9 ± 3.2 light particles per 100 ^{259}Md fission decays. This number is significant because at least one light particle for every 2-3 fission decays would be needed to explain the magnitude of the TKE deficit in the symmetric fission of ^{259}Md. Therefore, the hypothesis of ^{259}Md fission accompanied by significant light-particle emission cannot explain this energy deficit. Moreover, there is apparently no explanation for this problem available within the framework of current fission theory. New models of fission or modifications of existing theories seem necessary to explain this phenomenon while still maintaining consistency in predictions of fission properties for nuclides whose fission properties have already been determined. Further experimental work might also be performed to shed light on the problem. For example, the neutron multiplicity of ^{259}Md fission could be measured in order to determine if there is a significant amount of energy in the symmetric-fission mode distributed in fragment excitation.

We also redetermined the fission properties of ^{259}Md by measuring the kinetic energies of 333 pairs of coincident fission fragments. Thinner samples were obtained because of improvements in the chemistry used to purify ^{259}No. This narrowed the TKE distribution and lowered the fraction of events in the wings of the mass distribution. Individual points of TKE vs. fragment mass for all fission events are shown in Fig. 3. Figure 4 illustrates the symmetric mass division and the average TKE associated with each incremental group of masses. The important features of the energy distribution, namely the most probable TKE (190-200 MeV) and the average TKE at symmetry (~210 MeV) remain the same as measured before. This confirms that the TKE deficit problem is genuine and definitely unrelated to thick samples.

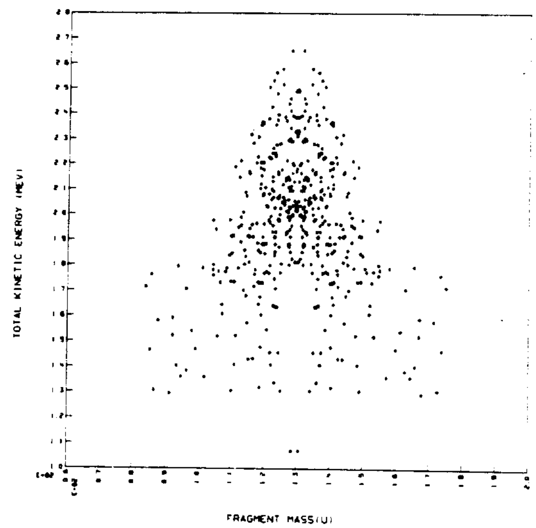

Fig. 3. Mass-TKE distribution from the spontaneous fission of ^{259}Md. Each fission coincident event provides two points which are reflected in the mass plane.

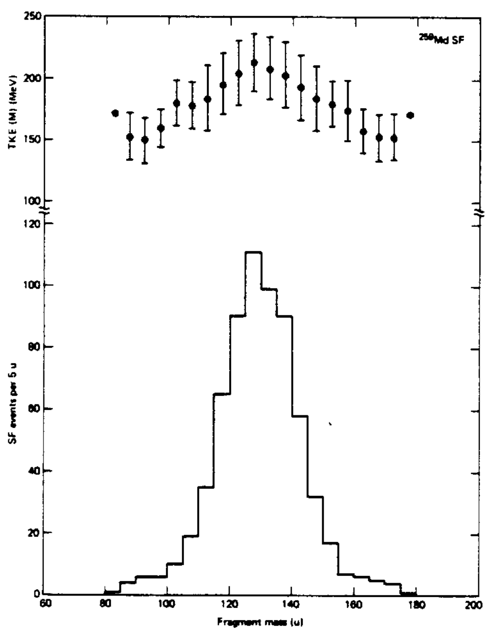

Fig. 4. Mass distribution from the spontaneous fission of ^{259}Md (bottom) and the average total kinetic energy (top) associated with each mass bin shown in the histogram below.

In sum, the low fragment energies at mass symmetry for the fission of ^{259}Md are unexplainable within the framework of current fission theory and are at considerable odds with the distribution between kinetic and potential energies found in the symmetric fission of ^{258}Fm and ^{259}Fm.

In addition to our investigation of the symmetric fission of ^{259}Md, we have made preliminary measurements of the light-particles emitted during the SF of ^{256}Fm and have just begun similar measurements with ^{257}Fm. Our purpose is to determine whether the potential energy available for this process is beginning to be reduced by the formation of colder, more spherical fragments at scission. The SF properties of ^{256}Fm and ^{257}Fm are clearly transitional between the "traditional" asymmetric, low-TKE fission of the lighter actinides, and the symmetric, high-TKE fission of ^{258}Fm and ^{259}Fm.

Much experimental and theoretical work has been performed over the past two decades to characterize the phenomenon of light-particle emission during fission (Ref. 4). The results of these studies suggest that ternary fission and binary fission are quite similar, except that some energy is expended in the formation and acceleration of the light particle which is predictably reflected in lower fragment energies and masses, and less neutron and γ-ray emission. Qualitatively, trajectory calculations indicate that at the scission point the fissioning nucleus in ternary fission is a little more elongated than in binary fission, and that the major fission fragments have achieved a considerable fraction of their final kinetic energies even prior to the snapping of the neck between them.

Within a given kind of fission, such as SF or fission induced by thermal neutrons, the rate of light-particle emission increases somewhat linearly with a variable related to the size of the nucleus, such as Z^2/A, which is the ratio of electrostatic to surface energy in the liquid-drop model of fission. This trend is illustrated in Fig. 5 for several SF nuclides and includes our preliminary measurement for ^{256}Fm. This trend might be expected, at least for those nuclides which exhibit the "normal" asymmetric mode of fission, since the fraction of available energy for fission which appears as fragment kinetic energy generally decreases with increasing nuclear size. A larger amount of potential energy is therefore available for the formation of light particles -- energy for producing the more elongated pre-scission nuclear shapes apparently necessary for the emission of light particles.

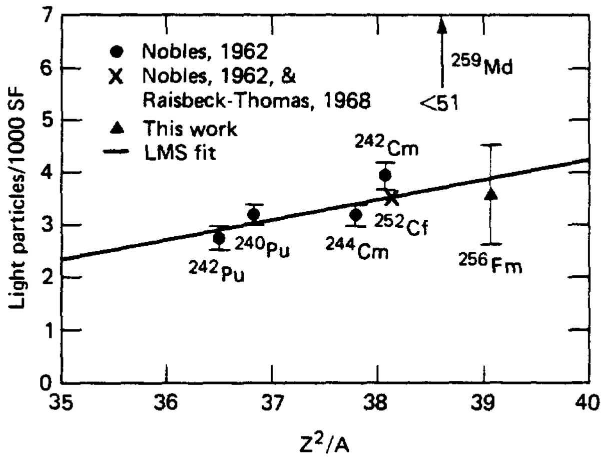

Fig. 5. The abundance of light particles emitted in the spontaneous fission of actinide nuclei as a function of the liquid-drop fissility parameter Z^2/A. The point for ^{256}Fm from our preliminary measurements indicates no deviation from these systematics.

This trend of increased light-particle emission with increasing nuclear size might be expected to continue on up through the heaviest actinides, if it weren't for the anomalous fission behavior of ^{258}Fm and ^{259}Fm. For these isotopes, most of the available energy for fission appears as fragment kinetic energies because the fragments themselves are relatively spherical and unexcited, owing their stability to their proximity to the doubly-magic nucleus ^{132}Sn. Since the emission of a light particle appears to cost the fissioning nucleus at least 25 MeV in potential energy, there is likely to be a large decrease in the light-particle emission rate for the SF of ^{258}Fm and ^{259}Fm due to a lack of deformation energy available for this process. The experimental proof of this supposition would require a major undertaking because of the short half lives of these nuclides (380 μs and 1.5 s, respectively). Therefore we have begun our study of nuclear shapes at scission with ^{256}Fm and ^{257}Fm where an anomaly in the rate of emission of light particles might first be detected.

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